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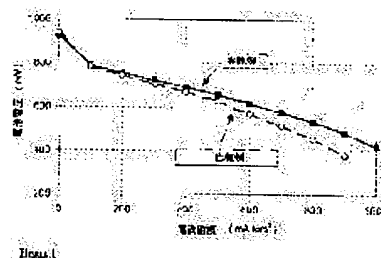
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(54) WATER-BASED CATALYST INK, AND USE OF THE SAME FOR MANUFACTURING BOARD COATED WITH CATALYST

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a means to coat a hydrophobic backing and another board with a water-based ink without a wettability problem.

SOLUTION: The water-based ink comprises (a) an electrocatalysis of 5 to 75 wt.% based on the ink weight, (b) a 10 to 75 wt.% ionomer solution based thereon, (c) water of 10 to 75 wt.% based thereon, (d) an organic solvent of 0 to 50 wt.% based thereon, and (e) a surfactant of 0.1 to 20 wt.% based thereon which has a vapor pressure of 1 to 600 Pa at ambient temperature.



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CLAIMS

[Claim(s)]

[Claim 1]

It is catalyst ink based on water, and this ink is the following. :

- (a) It is based on the weight of this ink and is the electrocatalysis of 5 - 75% of the weight of an amount.;
- (b) It is based on the weight of this ink and is the ionomer solution of 10 - 75% of the weight of an amount.;
- (c) It is based on the weight of this ink and is water of 10 - 75% of the weight of an amount.;
- (d) the weight of this ink -- being based -- organic solvent; of 0 - 50% of the weight of an amount -- and
- (e) It is based on the weight of this ink which has the vapor pressure of the range of 1-600 pascals in ambient temperature, and is the surfactant of 0.1 - 20% of the weight of an amount, Ink to contain.

[Claim 2]

Catalyst ink based on water according to claim 1 said whose vapor pressure of said surfactant in ambient temperature is 100-500Pa.

[Claim 3]

Catalyst ink based on water according to claim 2 chosen from the group which said surface active agent becomes from a fluorination wetting agent, the wetting agent based on tetramethyl-crepe-de-Chine-diol, the wetting agent based on soybean lectin, HOSUHO-amino-lipoid, and such mixture.

[Claim 4]

Catalyst ink based on water according to claim 3 whose concentration of said surfactant is for 0.1 - 10 % of the weight to the AUW of said catalyst ink.

[Claim 5]

It is the process for manufacturing the substrate by which catalyst coating was carried out equipped with the catalyst bed which carried out deposition a hydrophobic front face and on it, and these processes are the following processes. :

- (a) Process which offers the substrate which has a hydrophobic front face;
 - (b) The process in which it is the process which coats the hydrophobic front face of this substrate with catalyst ink, and this catalyst ink contains the electrocatalysis, an ionomer, a solvent, and a surfactant; be in a row.
 - (c) Process which dries the obtained substrate by which catalyst coating was carried out;
- This surfactant is a process which includes and has the vapor pressure of the range of 1-600Pa in ambient temperature here.

[Claim 6]

It is a process equipped with the catalyst bed by which deposition was carried out a hydrophobic gaseous diffusion layer and on it for manufacturing a gaseous diffusion electrode, and these processes are the following processes. :

- (a) process; in which it is the process which applies catalyst ink to a gaseous diffusion electrode, this catalyst ink becomes from the electrocatalysis, an ionomer, a solvent, and a surfactant here, and this surfactant has the vapor pressure of the range of 1-600Pa with ambient temperature -- and
 - (b) The process which dries this gaseous diffusion electrode,
- The process to include.

[Claim 7]

Before spreading of a catalyst bed, coating of said gaseous diffusion electrode is first carried out in a micro layer, and, subsequently they are desiccation and the process according to claim 6 by which calcining is carried out.

[Claim 8]

The process according to claim 5 from which the surfactant of the catalyst ink based on water is removed with the drying temperature of the range of 50-150 degrees C.

[Claim 9]

The process according to claim 7 to which calcining of said gaseous diffusion electrode is carried out at the temperature between 200 degrees C and 400 degrees C.

[Claim 10]

It is the approach of including the process which is the operation of said substrate which was manufactured according to claim 5, and by which catalyst coating was carried out, and incorporates the substrate with which this catalyst coating of this approach was carried out to the membrane electrode assembly.

[Claim 11]

It is the approach of including the process at which it is the operation of said gaseous diffusion electrode layer manufactured according to claim 6, and this approach builds this gaseous diffusion electrode into a membrane electrode assembly.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[Field of the Invention]

[0001]

This invention still more specifically relates to a polymer electrolyte membrane fuel cell (PEMFC) and a direct methanol fuel cell (DMFC) about the field of an electrochemical cell and a fuel cell.

[Background of the Invention]

[0002]

A fuel cell changes a fuel and an oxidizer into the electrical and electric equipment, heat, and water with two electrodes separated spatially. As the fuel, the gas which was rich in hydrogen or hydrogen may be used, and oxygen or air may be used as the oxidizer. The energy conversion process in a fuel cell is especially distinguished with the high effectiveness. For this reason, the fuel cell is gaining in importance on the migration application, the fixed application, and the portable way.

[0003]

Those small designs, power density, and since it is efficient, the polymer electrolyte membrane fuel cell (PEMFC) and the direct methanol fuel cell (it is the anomaly of DMFC and PEMFC and the direct output of this is carried out not with hydrogen but with a methanol) are suitable in order to use it as energy conversion equipment. The technique of a fuel cell is widely described by reference. For example, refer to nonpatent literature 1.

[0004]

The component of the base of a fuel cell is a membrane electrode assembly (MEA). This is equipped with the film which consists of a proton conductive polymer. This polymer ingredient may be used in order to be called "ionomer resin" and to form the ionomer film in this specification. In a fuel cell, the field of the opposite side of an electrolyte membrane touches the catalyst bed, and this carries out the catalyst of the electrochemical reaction. One catalyst bed of a membrane electrode assembly forms an anode, and the catalyst bed of another side forms a cathode. In an anode, hydrogen oxidizes, and oxygen reacts in a cathode with the proton which penetrates the ionomer film, and water and the electrical and electric equipment are produced.

[0005]

The film (it abbreviates to "CCM" hereafter) by which catalyst coating was carried out is equipped with a polymer electrolyte membrane, and this can prepare a catalytic activity layer in both sides. One side of those layers takes the gestalt of the anode which oxidizes hydrogen, and the 2nd layer takes the gestalt of the cathode which returns oxygen. Since CCM consists of three layers (an anode catalyst bed, the ionomer film, and cathode catalyst bed), it is often called "3 layer MEA."

[0006]

A "gaseous diffusion layer" ("GDL") (occasionally this is called a gaseous diffusion substrate or lining cloth) is arranged on an anode layer and a cathode layer, in order to establish electric contact at the same time it carries the gas reaction medium (hydrogen and air) to a catalytic activity layer. Although GDL usually consists of a substrate (for example, carbon fiber paper or carbon textile fabrics) based on carbon, these are very porosity and give good access to a catalyst bed to those reaction gases. furthermore -- while preventing intercepting this gaseous diffusion layer when the hole of these systems is flooded -- an anode -- humidification -- service water is offered and reaction water can be removed

from a cathode. In order to prevent flooding the hole of a gaseous diffusion layer, the hydrophobic polymer (for example, polytetrafluoroethylene (PTFE)) is immersed by GDL. GDL is convertible into the anode mold GDL or especially the cathode mold GDL depending on whether it forms in which MEA side.

[0007]

The carbon substrate with which GDL is manufactured shows a very coarse surface structure. So, GDL can be coated with a micro layer in order to raise contact to the catalyst bed of this fuel cell of GDL. A micro layer consists of conductive carbon black and mixture of a hydrophobic polymer (for example, polytetrafluoroethylene (PTFE)), and usually graduates a surface structure with a coarse carbon substrate.

[0008]

As mentioned above, a membrane electrode assembly is :core polymer electrolyte membrane, two catalyst beds, and two gaseous diffusion layers which consist of five layers. A polymer electrolyte membrane consists of a proton conductive polymer ingredient. These ingredients form the ionomer film. Preferably, the tetrafluoroethylene-fluoro vinyl-ether copolymer which has a sulfonic group is used. This ingredient is sold by for example, E.I.DuPont by the trade name of Nafion (trademark). However, other ionomer ingredients (for example, a sulfonation polyether ketone, an aryl ketone, or poly benzimidazole) which do not contain especially a fluorine may be used. The suitable ionomer ingredient is described by nonpatent literature 2. In order to use it for a fuel cell, generally these film has the thickness between 10 micrometers and 200 micrometers. Furthermore,; which is a hydrophilic property typically, however the advanced (advanced) ingredient of the front face of a polymer electrolyte membrane which has a hydrophobic front face are also well-known.

[0009]

In these anodes and a cathode catalyst bed, this carries out the catalyst of each reaction (oxidation of the hydrogen in an anode, and reduction of the oxygen in a cathode) including the electrocatalysis. Preferably, the metal of the platinum group of a periodic table is used as the catalytic activity component. In large ****, a support catalyst is used and the catalytic activity platinum metal is being fixed to the front face of a conductive support ingredient with the particle shape of nano size here. The mean particle diameter of this platinum metal is for about 1nm and 10nm. It has become clear that the carbon black which has the particle size of 10-100nm and high conductivity is also suitable as a support ingredient.

[0010]

The various methods of manufacturing a perfect membrane electrode assembly (MEA) are indicated. For example, coating of the electrolyte membrane is first carried out by the indispensable catalyst bed in both sides, and it may produce the catalyst coating film (CCM). In order to produce a membrane electrode assembly from there, GDL is arranged in the upper part of a catalyst bed, and needs to carry out a laminating. Or coating of the catalyst bed is first carried out to a gaseous diffusion layer, and it may produce the backing (CCB) which carried out catalyst coating. Subsequently, an electrolyte membrane is arranged between two backing which carried out catalyst coating, and it is established when the film contact between a total of three components puts heat and a pressure.

[0011]

Therefore, MEA may be manufactured by being manufactured by combining CCM (film by which catalyst coating was carried out), and two GDL(s) (an anode side and cathode side), or combining the ionomer film and two backing (CCB) which carried out catalyst coating by the anode and cathode side. In both cases, a five-layer MEA product is obtained. These two manufacture schemes may be combined if suitable.

[0012]

Catalyst ink may be used in order to produce CCB. Catalyst ink is pastiness matter which contains other components (for example, a hydrophobic polymer binder, a hole formation factor, etc.) the electrocatalysis, an ionomer, a solvent, and if needed. Subsequently, this ink is applied to the front face of a gaseous diffusion layer using a suitable technique, and is dried by heating. Thus, it is combined with the ionomer film and the prepared backing (CCB) by which catalyst coating was carried out can form a membrane electrode assembly.

[0013]

The solvent used in order to prepare ink usually contains water and an organic solvent. Depending on the amount of water, the ink (water forms most solvents used) based on water can be discriminated from ink (an organic solvent forms most).

[0014]

Use of catalyst ink is common knowledge at this contractor. For example, the patent reference 1 indicates the catalyst ink based on organic solvents (for example, propylene carbonate, ethylene carbonate, etc.) in most. However, since the ink based on water is not subordinate to the stringent Occupational Safety and Health regulation, most is desirable.

[0015]

In the patent reference 2, the approach of applying a catalyst to the electrode substrate which uses catalyst ink (Triton-X is included as Pt black, graphite, PTFE, water, and a surfactant) is proposed. It originates in the high boiling point and the low vapor pressure of Triton-X, and after a print process and a desiccation process, to apply a separate washing process and a rinse process is needed in order to remove a surfactant.

[0016]

The patent reference 3 indicates the catalyst bed prepared using polyvinyl alcohol (PVA). By this surface-active property of PVA offering suitable distribution of a between [the catalyst particles by which it is supported in a water solution], this monolayer structure acts in order to combine a carbon particle and a Nafion (trademark) ensemble, consequently the strong film is obtained from the low weight fraction of PVA. Though regrettable, PVA is the polymer ingredient surely disassembled by heating or washing (PVA is removed from a catalyst bed) by water.

[0017]

The patent reference 4 indicates the use of Triton-X 100 (surfactant) for preparing the ink containing carbon black and polytetrafluoroethylene (PTFE). This ink is used in order to coat a carbon cloth substrate. Then, this cloth is dried, PTFE is dissolved by heating for 30 minutes at 370 degrees C, a surfactant is disassembled at coincidence, and it removes.

[0018]

The patent reference 5 indicates the catalyst ink which contains water as the electrocatalysis, an ionomer, and a solvent. In addition to an ionomer, this ink does not contain the further organic component. The patent reference 6 indicates the catalyst ink containing the electrocatalysis, an ionomer, water, and an organic solvent, an organic solvent is at least one compound chosen from the straight chain dialcohol which has the flash point higher than 100 degrees C, and the amount of 1 - 50% of the weight of the range exists in ink to the weight of water here.

[0019]

When trying to coat a hydrophobic radical plate (for example, backing or a polymer film) with the ink based on the water of a hydrophilic property, when the coating needs to be applied with a big gestalt, the problem of serious wettability produces it especially. In order to deposit, and for there to be an inclination which forms an island (island), consequently to attain uniform coating, the coating process which some followed is required for the printed ink. This consumes time amount and is expensive.

[0020]

The means for coating hydrophobic backing and other substrates using the ink based on water based on the above-mentioned, without being accompanied by the problem of the above-mentioned wettability is needed in the field concerned. So, this invention does not need to apply some coating processes and conquers the property which crawls the water on the front face of hydrophobic of a substrate about the process for manufacturing the substrate by which catalyst coating was carried out using the catalyst ink based on water. This invention relates to offering catalyst ink suitable for this process again.

[Patent reference 1] U.S. Pat. No. 5,869,416 specification

[Patent reference 2] U.S. Pat. No. 4,229,490 specification

[Patent reference 3] U.S. Pat. No. 5,211,984 specification

[Patent reference 4] U.S. Pat. No. 6,127,059 specification

[Patent reference 5] the [Europe patent application public presentation] -- 0 731 No. 520 specification

[Patent reference 6] the [German country patent application public presentation] -- a 100 37 No. 074 specification

[Nonpatent literature 1] K. Kordes and G. Simader "Fuel Cells and its Applications" Germany, VCH

Verlag Chemie, Weinheim, 1996

[Nonpatent literature 2] O. Savadogo and "Journal of New Materials for Electrochemical Systems" I, 1998, p.47-66

[Description of the Invention]

[Problem(s) to be Solved by the Invention]

[0021]

The technical problem of this invention is offering the means for coating hydrophobic backing and other substrates using the ink based on water, without being accompanied by the problem of the above-mentioned wettability.

[Means for Solving the Problem]

[0022]

This invention offers the catalyst ink based on water, and this ink is the following. :

- (a) It is based on the weight of ink and is the electrocatalysis of 5 - 75% of the weight of an amount.;
- (b) It is based on the weight of ink and is the ionomer solution of 10 - 75% of the weight of an amount.;
- (c) It is based on the weight of ink and is water of 10 - 75% of the weight of an amount.;
- (d) the weight of ink -- being based -- organic solvent; of 0 - 50% of the weight of an amount -- and
- (e) It is based on the weight of ink which has the vapor pressure of the range of 1-600 pascals in ambient temperature, and is the surfactant of 0.1 - 20% of the weight of an amount,

It contains.

[0023]

In one operation gestalt, the above-mentioned vapor pressure of the above-mentioned surfactant in ambient temperature is 100-500Pa.

[0024]

It is chosen from the group which the above-mentioned surface active agent becomes from a fluorination wetting agent, the wetting agent based on tetramethyl-crepe-de-Chine-diol, the wetting agent based on soybean lectin, HOSUHO-amino-lipoid, and such mixture in one operation gestalt.

[0025]

In one operation gestalt, the concentration of the above-mentioned surfactant is for 0.1 - 10 % of the weight to the AUW of the above-mentioned catalyst ink.

[0026]

It is the process for manufacturing the substrate by which catalyst coating was carried out equipped with the catalyst bed which carried out deposition a hydrophobic front face and on it in another aspect of affairs, and this process is the following processes. :

- (a) Process which offers the substrate which has a hydrophobic front face;
- (b) The process in which it is the process which coats the hydrophobic front face of a substrate with catalyst ink, and catalyst ink contains the electrocatalysis, an ionomer, a solvent, and a surfactant; be in a row.

- (c) Process which dries the obtained substrate by which catalyst coating was carried out;

Including, this surfactant has the vapor pressure of the range of 1-600Pa in ambient temperature here.

[0027]

It is the process for manufacturing a gaseous diffusion electrode equipped with the catalyst bed by which deposition was carried out a hydrophobic gaseous diffusion layer and on it in one operation gestalt, and this process is the following processes. :

- (a) process; in which it is the process which applies catalyst ink to a gaseous diffusion electrode, this catalyst ink becomes from the electrocatalysis, an ionomer, a solvent, and a surfactant here, and this surfactant has the vapor pressure of the range of 1-600Pa with ambient temperature -- and
- (b) The process which dries this gaseous diffusion electrode,

It includes.

[0028]

In one operation gestalt, before spreading of a catalyst bed, first, coating of the above-mentioned gaseous diffusion electrode is carried out, and, subsequently it dries and calcines it in a micro layer.

[0029]

In one operation gestalt, the surfactant of the catalyst ink based on water is removed with the drying temperature of the range of 50-150 degrees C.

[0030]

In one operation gestalt, calcining of the above-mentioned gaseous diffusion electrode is carried out at the temperature between 200 degrees C and 400 degrees C.

[0031]

It is the approach of including the process which is the operation of the substrate which was manufactured according to the above-mentioned process, and by which catalyst coating was carried out [above-mentioned] in one aspect of affairs, and incorporates the substrate with which catalyst coating of this approach was carried out to the membrane electrode assembly.

[0032]

It is the approach of including the process at which it is the operation of the above-mentioned gaseous diffusion electrode layer manufactured according to the above-mentioned process in one aspect of affairs, and this approach builds a gaseous diffusion electrode into a membrane electrode assembly.

[0033]

This invention relates to those use for manufacture of the catalyst ink based on water, and the substrate by which catalyst coating was carried out. A catalyst bed uses the catalyst ink containing the electrocatalysis, an ionomer, and water based on water, and is applied to the hydrophobic front face of a substrate by this invention. This catalyst ink contains the very volatile surfactant which has the vapor pressure of the range of 1-600Pa in ambient temperature again. Use of this surfactant makes it possible to apply the ink based on water to the hydrophobic front face of various substrates (for example, a gaseous diffusion layer, the advanced ionomer film, and a polymer substrate). The deposition of coating needed is applied in a single coating phase, and is obtained, and the catalyst bed obtained shows an improvement of the engine performance resulting from the nonexistence of the surfactant which remains to a catalyst bed.

[0034]

(Summary of invention)

Although the following explanation is referred to together with an example in order to understand this invention better with other further profits and operation gestalten, the range of this invention is indicated by the attached claim.

[0035]

This invention relates to the catalyst ink based on water which may be used in order to manufacture the catalyst coating substrates for film fuel cells (for example, CCB, CCM, etc.). According to this invention, catalyst ink is produced by adding a surfactant in the catalyst ink based on water. The surfactant of high volatility which has the vapor pressure of 1-600Pa in ambient temperature (about 20-25 degrees C) is suitable for this purpose. Use of such a surfactant enables spreading of catalyst ink based on the water on the front face of hydrophobic of a substrate.

[0036]

The deposition ("deposition") of coating needed is attained in a single coating phase, and is obtained, and the catalyst bed obtained does not show reduction of the engine performance resulting from the surfactant which remains in the layer. In the drying temperature used between coating processes, a surfactant evaporates from ink, without remaining at all to a catalyst bed.

[0037]

According to one operation gestalt, this invention offers the catalyst ink based on water, and this ink contains the following. :

- (a) It is based on the weight of ink and is the electrocatalysis of 5 - 75% of the weight of an amount.;
- (b) It is based on the weight of ink and is the ionomer solution of 10 - 75% of the weight of an amount.;
- (c) It is based on the weight of ink and is water of 10 - 75% of the weight of an amount.;
- (d) the weight of ink -- being based -- organic solvent; of 0 - 50% of the weight of an amount -- and
- (e) It is based on the weight of ink which has the vapor pressure of the range of 1-600 pascals in ambient temperature, and is the surfactant of 0.1 - 20% of the weight of an amount.

[0038]

According to the 2nd operation gestalt, this invention offers the process for manufacturing the substrate by which catalyst coating was carried out equipped with the catalyst bed which carried out deposition a hydrophobic front face and on it, and this process includes the following processes. :

- (a) Process which offers the substrate which has a hydrophobic front face;

(b) The process in which it is the process which coats the hydrophobic front face of this substrate with catalyst ink, and this catalyst ink contains the electrocatalysis, an ionomer, a solvent, and a surfactant; be in a row.

(c) Process which dries the obtained substrate by which catalyst coating was carried out; Here, this surfactant is very volatility and has the vapor pressure of the range of 1-600Pa in ambient temperature.

[0039]

According to the 3rd operation gestalt, this invention offers the process equipped with the catalyst bed by which deposition was carried out a hydrophobic gaseous diffusion layer and on it for manufacturing a gaseous diffusion electrode. This process The process which coats the gaseous diffusion layer of hydrophobic phosphate with catalyst ink, and the process which dries the obtained gaseous diffusion electrode are included. Here this ink Consisting of the electrocatalysis, an ionomer, a solvent, and a surfactant, in altitude, this surfactant is volatility and has the vapor pressure of the range of 1-600Pa with ambient temperature here.

[0040]

This invention may be used combining the application technique which uses a coating process which is indicated by the Europe patent application number 02017238.3 concerning a share, the continuing leveling process, and a desiccation process if needed.

[Effect of the Invention]

[0041]

According to this invention, the means for coating hydrophobic backing and other substrates using the ink based on water, without being accompanied by the problem of the above-mentioned wettability is offered.

[Best Mode of Carrying Out the Invention]

[0042]

This invention relates to the ink containing a surfactant based on water. Especially this surfactant improves the wettability of this ink over the charge of a hydrophobic radical plate (for example, a polymer film, an advanced (advanced) ionomer film ingredient, or PTFE sinking-in backing).

[0043]

This invention is explained about a desirable operation gestalt here. Limiting [in / these operation gestalten are offered for assistance of an understanding of this invention and / any formats] this invention should not be meant, and it should not be interpreted such. All the alternatives that can become clear to this contractor in case this indication is read, an alteration object, and an equal object are contained in the intention of this invention, and within the limits.

[0044]

This invention is not meant as they are catalyst ink or a paper about a membrane electrode assembly. A reader refers to the suitable textbook about those themes about the further information if needed.

[0045]

This invention offers the ink containing the surfactant of the selected mold based on water. Especially this surfactant improves the wettability of this ink over the charges of a hydrophobic radical plate (for example, a polymer film, an advanced ionomer film ingredient, or PTFE sinking-in backing etc.).

[0046]

This invention relates to the catalyst ink containing the surfactant which has comparatively high vapor pressure. This vapor pressure makes removal of this surfactant easy between desiccation phases (this is preferably performed at the temperature of 50-150 degrees C). Consequently, the surfactant which remains in the printed electrode layer decreases more. This brings about next an improvement of the electric engine performance of MEA which used these ink and was manufactured. The surface active agent (for example, octyl-phenoxy-polyethoxylate (for example, Rohm & Haas Co., Triton X-100 manufactured as be alike)) which has contrastive very low vapor pressure (namely, less than 1Pa) remains in the electrode printed after the desiccation process, and inactivates a catalyst bed.

[0047]

a surfactant suitable in order to use it with this invention -- desirable -- ambient temperature (about 20-25 degrees C) -- it is -- the range of 1-600Pa -- more -- desirable -- the range of 100-500Pa -- and it has the vapor pressure between 200Pa and 400Pa most preferably. As an example of the surface active agent

of a suitable class, although nonionic, anionic, or a cationic surface active agent (for example, such mixture, such as a fluorination wetting agent (Fluorad system manufactured by 3M Co.), a wetting agent (Surfynol (trademark) system manufactured by Air Products and Chemicals Inc.) based on tetramethyl-crepe-de-Chine-diol, a wetting agent based on soybean lectin, or HOSUHO-amino-lipoid) is mentioned, it is not limited to these. The vapor pressure of the matter may be determined by the standard technique. Listing of such data is available again in "CRC Handbook of Chemistry and Physics", CRC Press LLC, and Boca Raton (USA).

[0048]

In addition to a surfactant, this catalyst ink contains water (preferably deionized water) as the electrocatalysis, ionomer resin, and a main solvent. the amount of the surfactant added -- the total presentation of catalyst ink -- being based -- 0.1 - 20% of the weight of the range -- and it is 0.1 - 10% of the weight of the range preferably. In addition to a surfactant, the ink based on water may contain the further organic solvent, an additive, a defoaming agent, a pore formation agent, a preservative, etc.

[0049]

Although the carbon black (for example, Pt/C or PtRu/C) supported as suitable electrocatalysis by the catalyst based on noble metals is mentioned in order to use it with this invention, it is not limited to these. However, the inorganic oxide containing an exotic powdered metal, noble-metals black and noble metals, or non-noble metals may be used.

[0050]

The desirable operation gestalt of this invention is related with the catalyst ink containing the surfactant of 0.1 - 20% of the weight of an amount based on water which has 5 - 75% of the weight of the electrocatalysis, 10 - 75% of the weight of an ionomer solution (based on an organic solvent based on water), 10 - 75% of the weight of deionized water, 0 - 50% of the weight of an organic solvent, and the vapor pressure of 1-600Pa. as a suitable organic solvent -- a glycol (for example, ethylene glycol --) A diethylene glycol, propylene glycol, dipropylene glycol, Butanediol and such mixture, alcohol (for example, one to C4 alcohol and such mixture), Ester (for example, ester and such mixture of one to C4 alcohol, and C1 - 4 carboxylic acids), Although the mixture of an aromatic series solvent (for example, benzene or toluene), aprotic bipolar solvents (for example, N-methyl pyrrolidone, ethylene carbonate, propylene carbonate, DMSO, etc.), and these matter is mentioned, it is not limited to these. Preferably, a glycol is used.

[0051]

The ionomer solution is marketed and contains the ionomer underwater or in an organic solvent typically. Generally, these contain 5 - 20% of ionomer. the mold of the electrocatalysis -- depending -- the weight ratio of the ionomer pair electrocatalysis -- usually -- 1:1-1:15 -- desirable -- 1:1-1:10 -- and it is 1:2-1:6 more preferably. An ionomer solution ensures that it dilutes with the further organic solvent water and if needed, and the ink obtained may be processed.

[0052]

This invention offers the process about manufacturing the substrate by which catalyst coating was carried out again, and this process includes the following processes. :

- (a) Process which offers the substrates (GDL, ionomer film, etc.) which have a hydrophobic front face;
- (b) The process which offers the catalyst ink based on water which consists of the electrocatalysis, an ionomer, a solvent, and a surfactant; in a row
- (c) Process which the hydrophobic front face of this substrate is coated [process] with this ink, and dries the substrate which is obtained, and by which catalyst coating was carried out;

Here, in altitude, this surfactant is volatility and has the vapor pressure of 1-600Pa in ambient temperature.

[0053]

This substrate is preferably chosen from the group which consists of an ingredient of a polymer film, the ionomer film, a carbon fiber, a carbon cloth, the carbon felt, or a paper matrix. This substrate may exist with the gestalt of the continuous roll as each sheet.

[0054]

In one partial operation gestalt, before spreading of a catalyst bed, coating of the hydrophobic front face is carried out, and, subsequently desiccation and "calcining (calcine)" may be carried out in a micro layer. Preferably, calcining is performed at the temperature between 200 degrees C and 400 degrees C.

A micro layer can consist of conductive carbon black and mixture of a hydrophobic polymer (for example, polytetrafluoroethylene (PTFE)), and can graduate a surface structure with a coarse carbon substrate.

[0055]

Here, since this invention has generally been indicated, this invention may be more easily understood with reference to the following examples. This example is not meant as limiting this invention, unless it is provided as instantiation and explains in full detail.

[Example]

[0056]

In order to manufacture a membrane electrode assembly (MEA) according to the process advocated, Surfynol (trademark) from Air Products and Chemicals Co. was used as a surfactant, and the catalyst ink based on water was prepared. Surfynol (trademark)420 has the vapor pressure of about 270Pa in ambient temperature (22 degrees C). The Nafion (trademark) water solution was used as a source of supply of the ionomer of catalyst ink. This ionomer was used with that acid gestalt. The following ink was prepared for the cathode and the anode. :

Catalyst ink for a cathode :

13.0g Electrocatalysis Elyst A 40 (40% Pt/C, OMG AG, Hanau)
50.0g Nafion (trademark) solution (11.4 % of the weight of underwater)
35.0g Water (deionization)
2.0g Surfynol420 (trademark)
(A total of 100.0g)

Catalyst ink for an anode :

14.0g PtRu-electrocatalysis (40% PtRu/C, OMG AG, Hanau)
48.0g Nafion (trademark) solution (11.4 % of the weight of underwater)
36.0g Water (deionization)
2.0g Surfynol420 (trademark)
(A total of 100.0g) .

[0057]

The catalyst ink for a cathode was prepared by mixing a catalyst with a Nafion (trademark) solution, water, and a surface active agent thoroughly with a high-speed stirring device. By screen printing, hydrophobic carbon fiber paper (HE paper from SGL-Carbon) was coated with this ink only in one step, and it was dried at two processes (75 degrees C for 3 minutes and 95 degrees C for 1 minute).

Homogeneity was coated with all the front faces of a gaseous diffusion layer by the catalyst bed. The obtained cathode gaseous diffusion electrode (GDE) had noble-metals loading of 0.4 mg Pt/cm². By the formula, the anode gaseous diffusion electrode was similarly manufactured by using the catalyst ink for an anode. Anode GDE had noble-metals loading of 0.3 mg Pt/cm² and 0.15 Ru/cm². As well as the case of Cathode GDE in order to attain uniform coating without formation of an "island" over all the front faces of a carbon fiber substrate, one step of coatings were required.

[0058]

Thus, Cathode GDE and Anode GDE which were manufactured were used, and MEA (membrane electrode assembly) was assembled. For this purpose, the ionomer film (Nafion(trademark)112;DuPont; 50 micrometers of thickness) by which coating is not carried out has been arranged between Anode GDE and Cathode GDE. Subsequently, the laminating of this assembly was carried out at the persistence time for 1 minute under the pressure of 20bar, and the temperature of 150 degrees C.

[0059]

(Example of a comparison)

Catalyst ink was prepared by the formula like what was indicated by the above-mentioned example for the comparison. However, Triton X-100 (this is extensively used in fuel cell industry) was used as a surfactant instead of Surfynol (trademark). This surfactant has the vapor pressure of less than 1Pa in ambient temperature (22 degrees C). Therefore, this surfactant has volatility much lower than Surfynol (trademark). It prepared by the formula like what is indicated by the example which precedes a membrane electrode assembly using the catalyst ink based on two kinds of water.

[0060]

(Electrochemical trial)

It measured by inserting the membrane electrode assembly from an example and the example of a comparison in the PEMFC single trial cell which has the activity area of 2 50cm, and using reformat for these electrochemical engine performance as a fuel gas about an anode, and using air about the oxidizer in a cathode. The anode gaseous mixture contained 45 volume % H₂, 31 volume % N₂, 21 volume % CO₂, and the 50 volume ppm CO with the further air bleed of 3 volume %. Air was supplied to the cathode of a trial cell. The temperature of a cell was adjusted at 70 degrees C. It is 80 degrees C about damping of an anode, and the cathode was damped at 55 degrees C. The actuation gas pressure force was set as 1bar (absolute pressure). The stoichiometry of a reactant was adjusted [gas / anode] to 2.0 about 1.1 and a cathode gas.

[0061]

The electrical potential differences of the cell measured about the selected current density are enumerated to Table 1, and are shown in drawing 1 . It is proved [results / these] clearly that MEA manufactured by using the catalyst ink by this invention migrates to all current density range, and has the electrochemical engine performance improved considerably. The difference in the electrical potential difference of a cell also increases as shown in drawing and current density increases, when the electrical potential difference of the cell of an example is compared with the example of a comparison.

[0062]

A surfactant Surfynol(trademark) 420 and Triton X-100 have different vapor pressure in ambient temperature. The vapor pressure of Surfynol (trademark) is more nearly intentionally [than the vapor pressure of Triton X-100] high. Therefore, Surfynol (trademark) evaporates easily from catalyst ink in the desiccation conditions applied. A surfactant is not substantially contained in the catalyst bed left behind, consequently it has the very good electrochemical engine performance in it. By contrast, including the surfactant of amount with the still most catalyst bed which Triton X-100 carried out chisel evaporation slowly from catalyst ink, consequently was dried, this is considered to intercept the active site of the electrocatalysis partially, therefore brings about the scarcer engine performance.

[0063]

Table 1: Electrical potential difference of cell measured to current density as which membrane electrode assembly from example and example of comparison was chosen (mV)

[0064]

[Table 1]

電流密度	[mA/cm ²]	100	500	900
実施例	[mV]	791	658	484
比較例	[mV]	784	627	381

[Availability on industry]

[0065]

This invention has the availability on industry in the field of an electrochemical cell and a fuel cell, and still more specifically has the availability on industry in a polymer electrolyte membrane fuel cell (PEMFC) and a direct methanol fuel cell (DMFC).

[Brief Description of the Drawings]

[0066]

[Drawing 1] Drawing 1 shows the U/I performance curve (cell voltage pair current density) about MEA manufactured by using the catalyst ink (example 1) by this invention in comparison with MEA produced by using conventional catalyst ink (example of a comparison).

[Translation done.]